## N95-11069

# variation of stratospheric $\mathrm{no_2}$ during the solar eclipse

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#### ABSTRACT

The numerical modeling of the changes of the concentration of trace gases in the atmosphere during the aclipse shows that the NO<sub>2</sub> total content in the vertical column increases approximately by 80 %. The first observations of the NO<sub>2</sub> total content during the eclipse of 1981 have given 60±20%. In the observatios of 1990 the more precise methods and instruments for stratospheric NO<sub>2</sub> measurements were used. The surface ozone, NO and NO<sub>2</sub> concentrations were under control. The results of the observations give the increasing of the stratospheric NO<sub>2</sub> during the eclipse by 55  $\pm$  6%. The maximum increasing of the NO<sub>2</sub> content is observed at the moment of the maximum phase.

#### 1. INTRODUCTION

The variations of the trace concentration in the atmosphere during the solar eclipse was investigated with the help of the numerical photochemical modeling. The first estimations of the variability of the ozone estimations of the variability of the ozone concentration in the stratosphere were made by Hunt (1965) with only Chapman system of reactions taken into consideration. Herman (1979), Wuebbles and Chang (1979) on the base of the more complete system of photochemical intractions had investigated the behavior of the other constituents of the atmosphere. Later their calculations were specified by Gruzdev and Elansky (1982) with the help of the photochemical constituents of the model, which took into account the variation of the spectral composition of the sunlight and the temperature of the atmosphere during the total eclipse. In particular they have obtained that the total NO<sub>2</sub> amount in the vertical column had increased approximately by 84% but the total ozone practically had not changed.

The experiments carried out during the solar eclipse 31 July 1981 confirmed on the whole the results of calculations (Elansky et al., 1982, Elansky et al., 1983). The measurements of the  $NO_2$  total amount were based on the absorption of the direct sunlight in the  $NO_2$  absorption band and

outside of it at 4 wavelengths: 442.0; 441.3; 439.0 and 437.8 nm.

During the eclipse the increase of the  $NO_2$  amount in the atmosphere had been observed. The most increasing, equal to  $60\pm20\%$ , was registered during the first 10 minutes after passing the maximum phase. Such variations of the  $NO_2$  amount agree with the results of the calculations, if to take into account that the eclipse at the observation point was not total and the durations of its influence on  $NO_2$  was shorter than in the model, as it began before the sunrise.

But the next cause prompted us to carry out the repeated measurements of the NO<sub>2</sub> content during the eclipse. Firstly, there was the big error of the measurements based on the absorption of the direct sunlight. Secondly, during the measurements there was no control of the surface NO<sub>2</sub> concentration. Though the probability of the essential increasing of the NO<sub>2</sub> concentration near the surface under the background conditions of alpine is small, nevertheless the influence of the local sources of NO<sub>2</sub> or its long-way transport cannot be excluded completely. Some cases of increasing of the NO<sub>2</sub> concentration up to 3-5 ppb with the duration up to several hours are being registered by us episodically at the scientific station "Kislovodsk" (North Caucasus, 2070 m a.s.l.).

#### 2. OBSERVATIONS

The observations of NO<sub>2</sub> during the eclipse 22 July 1990 were carried out at the village Kondratievo (60.62 N, 28.17 E) situated 10 km away from the Baltic seaside in the rural region.

The maximum phase here was 0.995, which corresponded to the covering of 99,7% of the solar disk, and was registered at 1 hour 52 min 15 s GMT. Solar zenith angle was at this moment 87,0°. The eclipse had began before sunrise  $(\Theta_0=92.4^\circ)$ , the altitude of the earth's shadow was at that moment 7 km (i.e. below the main layer of NO<sub>2</sub>). Thus the parameters of the eclipse were near to optimal for the observations of the variations of stratospheric NO<sub>2</sub>.

According to the observation data (Analytical review, 1988) the mean  $NO_2$  concentration in the surface air on the west frontier of Russia in summer is 3-4 ppb. Its high values have been explained by the polluted air transport from the West and Central Europe. Apparently during the long-way transport the higher  $NO_2$  concentrations can occupy a rather thick atmospheric layer and influence the total its amount in the vertical column. The local sources of the nitrogen oxides ( $NO_X = NO + NO_2$ ) can cause considerable variations of the  $NO_2$  surface concentration as well. But, as a rule, their action is restricted by the thin surface layer of the air and influence on the total  $NO_2$  content, is insignificant.

The most part of the nitrogen oxides injects into the atmosphere in the form of NO. During the daytime, owing to the action of the photochemical cycle:

$$NO + O_3 \longrightarrow NO_2 + O_2$$
  
 $NO_2 + hv \longrightarrow NO + O$   
 $O + O_2 + M \longrightarrow O_3 + M$ 

the photochemical equilibrium takes place between NO, NO<sub>2</sub>, O<sub>3</sub>. By controlling the content of the trace gases in the surface layer and the intensity of solar radiation we can find the deviations of the concentrations from the equilibrium values and make the definite conclusions about the causes of the NO<sub>2</sub> variability.

Some influence on the gas composition of the surface layer of the atmosphere can exert the solar eclipse itself. The results of the observations have showed that the ozone concentration decreased during the eclipse (Britaev et.al., 1983). Probably some definite effect can take place and for nitrogen oxides. But under the condition when the eclipse coincides with the surrise such effect in the surface layer cannot be significant.

The measurements of the ozone concentration during the eclipse 22 July 1990 were carried out with the help of gas-analyzer Dasibi 1008-AH. The precision of the measurements is 1-2 ppb. The time of the respond is 50 s.

The measurements of the NO and NO<sub>2</sub> concentration were carried out with the chemoluminescent gas-analyzer Antechnika AC-30M. The precision of the measurements is 1 ppb. The time of the respond is 50 s.

Both instruments have been working continuously since 21 to 25 July 1990. The air was sampled on the level of 2 m above the ground.

The measurements of the NO<sub>2</sub> content in the stratosphere were based on the absorption of the scattered at the zenith sunlight in the visible region of the spectrum 430-450 nm. The spectrophotometer with the diffraction grating was used. The spectral resolution of the device is 1 nm. The spectra were registered with the step of 0.02 nm. The measurements were carried out at the solar zenith angles 78-96°. The spectral method proposed by Solomon et al.(1987) was applied. It makes possible to find the NO<sub>2</sub> slant abundance along the beam in the atmosphere and then retrieve the NO<sub>2</sub> content in the vertical column according

to the given optical parameters of the atmosphere.

### 3. DISCUSSION

The hourly means of the ozone and nitrogen oxides concentrations in the near-surface air since 21 to 25 July 1990 are given in Fig.1. Their values and daily variations are typical for the rural area (see, for example, Colbeck, 1989). Averaged over the observation period values of concentration are 20 pph for O<sub>2</sub> and 1.5 ppb for NO<sub>2</sub>.

20 ppb for O<sub>3</sub> and 1,5 ppb for NO<sub>x</sub>.

During 22 July the NO<sub>2</sub> concentration varied insignificantly around the level of 1 ppb. During the night from 21 to 22 July there was anticyclonic weather characterized by the absence of the clouds and wind and the presence of the near-surface temperature inversion. The O<sub>3</sub> concentration under such conditions fell down to the very small values.

The observation data and the analysis of the weather maps showed that the wet air mass, which

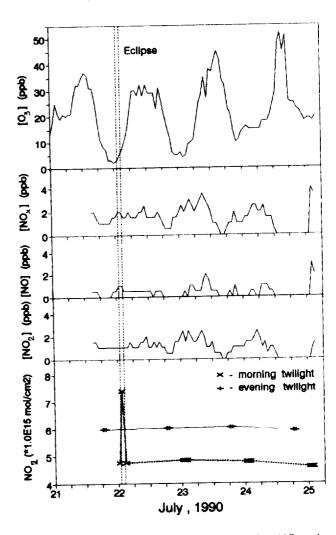


Fig. 1. Variations of surface ozone, NO<sub>x</sub>, NO and NO<sub>2</sub> concentrations and NO<sub>2</sub> vertical column amount at Kondratyevo for 21-25 July 1990.

occupied the area of observation on 22 July had come from the North Atlantic through the relatively clear central regions of Normay and Sweden.

In details the variation of the trace gases concentrations during the eclipse in comparison with the same period on 23 July is shown in Fig.2. The influence of the local sources of  $NO_{\chi}$  (the motor vehicles ) reveals itself as episodes of increasing of the NO concentration, which do not transform into  $NO_2$  because of the short time of its stay in the atmosphere and small ozone concentration.

There was detected no peculiarities in the behavior of NO and NO $_2$  connected with the eclipse. It is possible that the effect of the eclipse reveals itself in the decreasing of the O $_3$  concentration just after the passing of the maximum phase. The delay of the O $_3$  concentration minimum was 20 min.

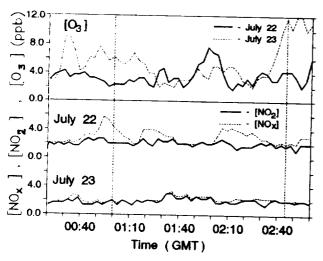


Fig. 2. Variations of surface ozone, NO<sub>X</sub> and NO<sub>2</sub> concentrations during sunrises of 22 and 23 July 1990. Vertical lines show the moments of maximum and end eclipse.

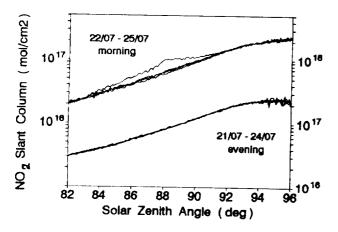


Fig. 3. The NO<sub>2</sub> slant amount on 21-24 July 1990 (left-morning, right-evening).

The analysis of the trace gases concentration data brings us to the conclusion, that the  $NO_2$  content in the lower layer of the atmosphere have corresponded to the background values (1 ppb) and there was no its variations, which could reveal themselves significantly in the  $NO_2$  content in the vertical column.

The measurements of the NO<sub>2</sub> slant amount were carried out since the evening of 21 July trough the morning of 25 July at sunrise and sunset (Fig. 3) The morning and evening NO<sub>2</sub> content values have the characteristic difference which is connected with the existence of the daily variations of the NO<sub>2</sub> content in the atmosphere

of the NO<sub>2</sub> content in the atmosphere.

There is a good stability of the NO<sub>2</sub> amount during this period. Only on 25 July the NO<sub>2</sub> content has changed a little because of the replacement of the air mass. On the background of the nearly constant values of the NO<sub>2</sub> content the effect provoked by the eclipse is distinctly marked out.

In accord with the numerical calculations of the radiation transport in the atmosphere the present method gives the NO<sub>2</sub> slant column abundance approximately down to the lower border of its stratospheric layer which is situated at the altitude 14-16 km. For the given latitude and the time of the year "Enhancement factor" which determines the ratio of the NO<sub>2</sub> slant column abundance to its content in the vertical column is 20,4 at  $\theta=90^{\circ}$ .

The calculated values of the stratospheric NO<sub>2</sub> content in the vertical column are given in Fig. 4 for 22 July and for the whole period in Fig. 1. The maximum increasing of the NO<sub>2</sub> content during the eclipse was 55±6% and is observed at the moment of the maximum phase. Calculating the error of the NO<sub>2</sub> measurements we have taken into account, that during the eclipse the structure of the solar spectrum and the value of the Ring-effect have been altering. But the analysis of the residual dispersion of the signal (Solomon et.al., 1987) have showed that the eclipse hadn't practically altered in comparison with usual days its

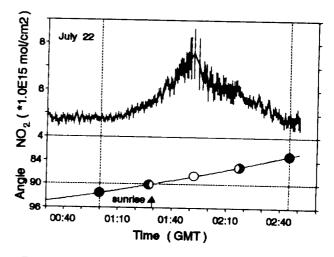


Fig. 4. The NO<sub>2</sub> vertical column amount for morning of 22 July 1990.

dependence on the intensity and its normal distribution along the spectrum. This is why it can be considered that the influence of the spectrum deformation on the  $NO_2$  measurements is small.

Table. Parameters of the solar eclipses: maximum phase ( $\phi$  max), duration (T), solar zenith angle at the eclipse's beginning ( $\theta_0$ ) and its maximum phase ( $\theta_{max}$ ), maximum change of the NO<sub>2</sub> content ( $\Delta$  NO<sub>2</sub>) and its delay about it maximum phase (T<sub>min</sub>).

φ max		<b>9</b> <sub>0</sub>	e max	T	ΔNO <sub>2</sub>	T <sub>min</sub>
1991	0.925	95.8	85.4	104	60±20	3
1990	0.997	92.4	87.9	114	55±6	0
Model	1.0	30.0	30.0	150	84	2

The increasing of the NO<sub>2</sub> content differs from the model estimations (Gruzdev and Elansky, 1982), but coincides within the limits of the errors with the change of the NO<sub>2</sub> total content above 2700 m (60±20%) after the observations of 31 July 1981 (Elansky and Arabov, 1982). As the conditions of the observations and the eclipses of 1981 and 1990 are very similar (see Table) we can consider that the results of 1990 are on the whole the specifying of the observation data of 1981. The model calculations didn't contradict to the results of the observations, as they were got for the total solar eclipse of the more duration.

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